

Laser induced ultrafast demagnetization: an *ab-initio* perspective

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Time-dependent density functional theory is implemented in an all electron solid-state code for the case of fully non-collinear spins. We use this to study laser induced demagnetization in Fe, Co and Ni. It is shown that this demagnetization is a two-step process: excitation of a fraction of electrons followed by spin-flip transitions of the remaining localized electrons. These results successfully explain several experimental features such as the time-lag between the start of the pulse and demagnetization and spin-flip excitations dominating the physics. We further show that it is possible to control the moment loss by tunable laser parameters like frequency, duration and intensity.

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Manipulation of electrons by femtosecond (fs) laser pulses opens up the vast and largely unexplored physical landscape of ultra-short time scales. One possibility in this landscape is to use electronic spins, which can be optically manipulated (flipped) using lasers to store data as binary bits. The advantage of such a technique would be a increase in the speed of data storage by orders of magnitude. Ultrafast light induced demagnetization[1] was demonstrated in the 90s[2], where demagnetization times (in Ni) faster than a few pico-seconds were achieved using intense laser pulses[3]. Recently, these demagnetization times have been measured down to a few hundred femtoseconds, owing to advances made in the refinement of pump-probe and other experiments[2, 4–11]. However, we are still far from achieving sufficiently controlled manipulation[10] of spins required for the production of useful devices. One of the reasons behind this is the lack of full understanding of the phenomena leading to laser induced demagnetization.

There have been a number of attempts at explaining this laser induced loss of moment: Combined action of spin-orbit coupling and interaction between spins and laser photons[12]. Super-diffusive spin transport where excited electrons carry spin with them from one part of the sample to another[13, 14]. Elliott-Yafet mechanism where electron-phonon or electron-impurity mediated spin-flip is the major contributor[15, 16]. Transfer of spin angular momentum to the lattice[17]. All these studies have in common that they describe the dynamics of the *excited electrons* using parameterized model Hamiltonians[18].

Time-dependent density functional theory (TDDFT)[19], which extends density functional theory into the time domain, is a formally exact method for describing the real-time dynamics of electrons under the influence of an external field such as the vector potential of the applied laser pulse. The advantage of such a technique is clear: it does not require any empirical parameter, is fully *ab-initio* and not only linear, but

also highly non-linear processes are a natural part of the simulation.

In the present work we use spin-resolved TDDFT to study the process of laser induced demagnetization. Magnetic non-collinearity can be a major contributor in the loss of moment so in order not to exclude such effects we extended TDDFT to the fully non-collinear case. We have further implemented this fully non-collinear magnetic time propagation for periodic systems in an all-electron code. Several bulk systems (Fe, Co and Ni) are studied using this code in order to explore various possible demagnetization scenarios. With optimal control of spins in mind we have also explored the effect of various tunable laser parameters of a laser pulse on the process of demagnetization.

Our analysis shows that the demagnetization occurs as a two-step process, where first the electrons make transitions to excited states followed by spin-orbit mediated spin-flip transitions. The interesting point about these results is that it is *not the excited delocalized electrons* which make these spin-flip transitions leading to the demagnetization of the material, but rather the remaining localized electrons. We note that while the non-collinearity of the spins is essential, long range coherent process like magnons do not play a significant role in the demagnetization in bulk systems for the time scales studied in the present work. These results explain several experimental indications like time-lag between the start of the pulse and moment loss[2, 5, 20] as well as spin-flip excitations dominating the initial demagnetization process[5, 14]. It is further demonstrated that the spins can be controlled using easily tunable laser parameters, for example the magnitude of the loss in moment (in Ni) can be controlled by changing intensity, frequency and/or duration of the laser pulse.

The Runge-Gross theorem[19] establishes that the time-dependent external potential is a unique functional of the time-dependent density, given the initial state. Based on this theorem, a system of non-interacting par-

ticles can be chosen such that the density of this non-interacting system is equal to that of the interacting system *for all times*. The wave function of this non-interacting system is represented as a Slater determinant of single-particle orbitals. In what follows we shall employ the non-collinear spin-dependent version of these theorems. Then the time-dependent Kohn-Sham (KS) orbitals are Pauli spinors determined by the equations:

$$i \frac{\partial \psi_j(\mathbf{r}, t)}{\partial t} = \left[\frac{1}{2} \left(-i\nabla + \frac{1}{c} \mathbf{A}_{\text{ext}}(t) \right)^2 + v_s(\mathbf{r}, t) \right. \\ \left. + \frac{1}{2c} \vec{\sigma} \cdot \mathbf{B}_s(\mathbf{r}, t) + \frac{1}{4c^2} \vec{\sigma} \cdot (\nabla v_s(\mathbf{r}, t) \times i\nabla) \right] \psi_j(\mathbf{r}, t) \quad (1)$$

where $\mathbf{A}_{\text{ext}}(t)$ is a vector potential representing the applied laser field, and σ are the Pauli matrices. The KS effective potential $v_s(\mathbf{r}, t) = v_{\text{ext}}(\mathbf{r}, t) + v_H(\mathbf{r}, t) + v_{\text{xc}}(\mathbf{r}, t)$ is decomposed into the external potential v_{ext} , the classical electrostatic Hartree potential v_H and the exchange-correlation potential v_{xc} . Similarly the KS magnetic field is written as $\mathbf{B}_s(\mathbf{r}, t) = \mathbf{B}_{\text{ext}}(t) + \mathbf{B}_{\text{xc}}(\mathbf{r}, t)$ where $\mathbf{B}_{\text{ext}}(t)$ is the magnetic field of the applied laser pulse plus possibly an additional magnetic field and $\mathbf{B}_{\text{xc}}(\mathbf{r}, t)$ is the XC magnetic field. The final term of Eq. (1) is the spin-orbit coupling term. Since the wavelength of the applied laser in the present work is much greater than the size of a unit cell we apply the dipole approximation and hence disregard the spatial dependence of the vector potential.

The exchange-correlation potential has a functional dependence on the density and the magnetization density of the system at the current and all previous times. Hence it includes the information about the whole history of this time propagation. Knowledge of this functional would solve all time-dependent (externally driven) interacting problems. In practice however, the exchange-correlation potential is always approximated. In the present work we use the adiabatic local spin density approximation (ALSDA)[21]. Using the method outlined above, various extended magnetic systems are studied[22] using the full-potential linearized augmented plane wave (FP-LAPW) method [23], implemented within the Elk code [24].

Presented in the middle panel of Fig. 1 are the magnetic moments of bulk Fe, Co and Ni as a function of time under the influence of an intense laser pulse[25]. In all cases, demagnetization is observed—the largest loss of moment is for Ni (43%) and the smallest for Fe (12%). We observe that in all three cases the systems do not become non-collinear in the sense that the loss of moment in the z -direction would be gained in the x or y -direction. It may be argued that since these calculations are performed using a single atom unit cell with periodic boundary conditions, it is premature to make any conclusions about the contribution of non-collinearity to the loss of moment. Hence we have studied the effect of the same laser pulse on a Ni unit-cell 4 times as large in size. So as not to bias our calculations towards collinear-

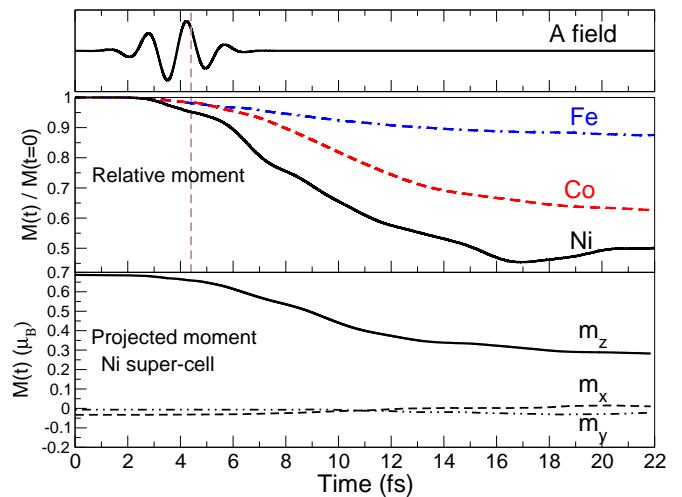


FIG. 1: (Color online) Top panel: $\mathbf{A}(t)$ of the laser pulse[25]. Middle panel: relative magnetic moment for Fe, Co and Ni. Lower panel: x , y and z -projected magnetic moment per atom (in Bohr magneton) for a super-cell of Ni. All times are in femtoseconds

ity we started (at 0 fs) from a random configuration of spins with respect to one other in this super-cell and the results for the moment (per Ni atom) projected in x , y and z -directions are presented in the lower panel of Fig. 1. While it is essential that non-collinearity is included in the calculation (due to the presence of the spin-orbit coupling term in Eq. (1)), we find that long-range non-collinearity, like the relative alignment of moments between atomic sites, does not play a significant role. This is mostly due to the fact that for the small time scales of interest in the present work one does not expect low energy non-collinear processes like magnons or spin-waves to dominate. Interestingly we note that the dimensionality plays an important role in this: for lower-dimensional systems like mono-layers and super-structures, inter-site non-collinearity of spins was found to be important.

A feature of the demagnetization process in all three cases (Fe, Co and Ni) is a time lag of ~ 5 fs between the start of the laser pulse and the beginning of the loss in the magnetic moment. Such a time lag has been noted in almost all experiments with the actual value of the time-lag depending upon the laser pulse used[2, 5, 20, 26, 27]. The questions now arise what is the origin of this time lag and what causes the demagnetization? To understand this, we plot in Fig. 2 two contributions to the moment for Fe and Ni. They are (a) from the electrons which under the influence of intense laser pulse make a transition to excited states and become delocalized and (b) from the remaining electrons which are localized close to the nuclei. During the first ~ 5 fs the electrons, carrying their spins, make transitions to excited states. This leads to an increase in the total moment associated with the excited electrons and to a lowering of the moment

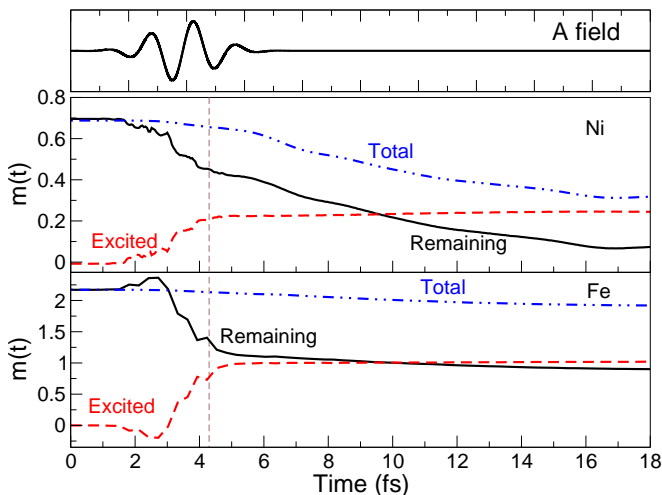


FIG. 2: (Color online) Top panel: $A(t)$ of the laser pulse[25]. Middle panel: total magnetic moment per Ni atom, magnetic moment per Ni atom coming from excited and remaining electrons. Lower panel: same as middle panel but for Fe. All moments are in Bohr magneton and times in femtoseconds.

coming from the remaining localized electrons such that the sum of the two moments stays almost constant. After ~ 5 fs the moment of the excited delocalized electrons stays almost constant while some of the remaining localized electrons make spin-flip transitions leading to a loss in the total moment. Hence the demagnetization mechanism is clearly a two step process: (1) during the first ~ 5 fs a fraction of the electrons becomes delocalized by making transitions to the excited states (2) this is followed by the remaining localized electrons making spin-flip transitions. The major factor responsible for these spin-flip transitions of the localized electrons is the spin-orbit coupling[28, 29] term in Eq. (1). To confirm this fact we also performed similar calculations in which the spin-orbit coupling term was set to zero, and find no such demagnetization.

This two-step process for demagnetization provides a clear explanation of the time-lag and is in line with experimental work which indicates fast charge dynamics followed by slower spin-dynamics[2, 26], and also with experiments which show a difference in the dynamics of spins and orbital angular momentum[30]. These results are also consistent with the idea that the indirect effect of spin-orbit coupling is slower than the direct absorption of light. These results also agree with the experimental results which show that it is indeed the spin-flip excitations which lead to demagnetization in the first picosecond and the non-collinear processes only start to have a substantial contribution much later[5, 14].

In the present work periodic boundary conditions are used and the excited electrons can not move away (diffusive mechanisms like proposed in Refs. [11, 13] are thus ignored). In a more realistic scenario such a dif-

fusion of electrons will lead to further demagnetization. Such a process would also contribute in the absence of any spin-orbit coupling and we expect demagnetization to be more than what is seen here. Other phenomena like lattice vibrations or radiative effects are not expected to contribute substantially for the time scales relevant to the present work.

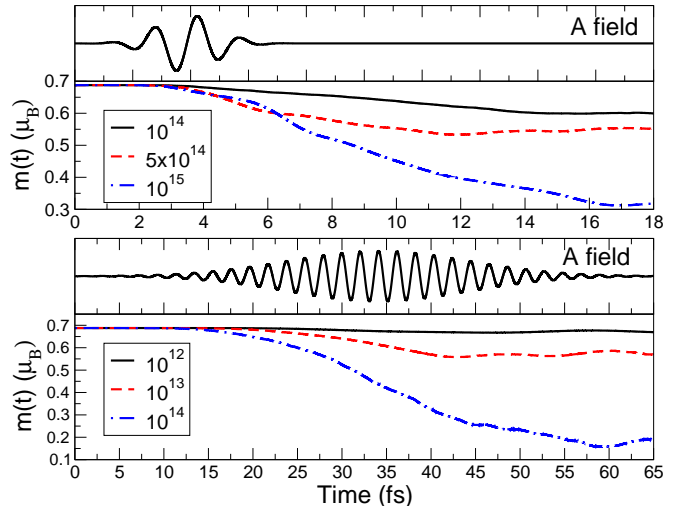


FIG. 3: (Color online) Top panel: $A(t)$ of the laser pulse of duration 6 fs. Second panel: total magnetic moment per Ni atom under the influence of laser pulses with three different peak intensities (see Ref. 31). Third panel: $A(t)$ of the laser pulse of duration 60 fs. Lower panel: total magnetic moment per Ni atom under the influence of laser pulses with three different peak intensities (see Ref. 32). All moments are in Bohr magneton and times are in femtoseconds.

The above analysis scrutinizes the process of demagnetization. What is most important for future technological applications is not just this knowledge but also the means to manipulate spins in a controlled manner. We now show the effect of the three easily tunable parameters of a laser pulse; intensity, frequency and duration on demagnetization. In the upper panel of Fig. 3 is shown the magnetic moment for Ni as a function of the peak intensity[31]. The effect of intensity on the demagnetization is dramatic. For a pulse of peak intensity 10^{15} W/cm² 35% of the moment is lost after 20 fs while only 14% loss is observed for a pulse of peak intensity 10^{14} W/cm². It is important to mention that at these intensities the response of the system is far from linear and non-linear effects are predominant.

In all these studies, a very short laser pulse of 6 fs was applied[31]. Most experiments are currently limited to using much longer laser pulses. In the lower panel of Fig. 3, we show the effect of such a long pulse of varying intensities[32]. Again we find that demagnetization increases with increasing intensity. For longer duration pulses, a higher demagnetization, with 71% loss in moment induced by the pulse of peak intensity 10^{14} W/cm²,

is observed. Also a clear indication from Fig. 3 is that for longer duration pulses, a lower intensity is sufficient to obtain a large demagnetization. We note that the time-lag between the start of the pulse and demagnetization depends upon the duration and frequency of the pulse and increases to ~ 15 fs on going from a pulse of duration 6 fs to 60 fs. Here we also observe that the physics of moment loss remains exactly the same i.e. a two-step demagnetization process with a time-lag. Hence the use of ultra short laser pulses in the present work is a matter of convenience for obtaining results within reasonable computer time by not having to propagate for very long times. An added benefit of short time scale pulses is also that we can clearly separate the spin-dependent phenomena from charge only excitations, which gets mixed with each other on large time scales.

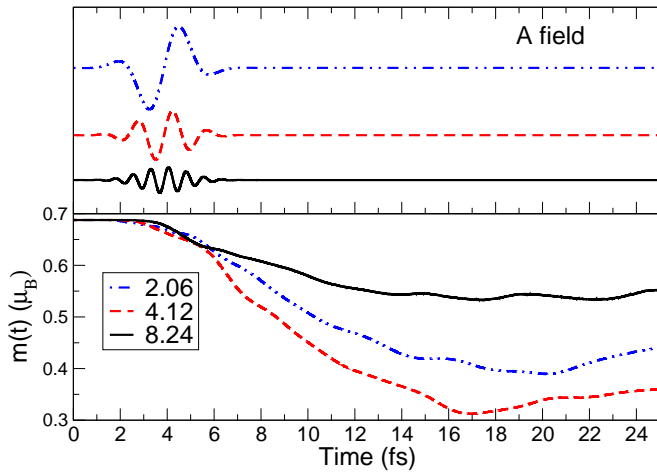


FIG. 4: (Color online) Top panel: $\mathbf{A}(t)$ of the laser pulses of peak intensity 10^{15} W/cm 2 and varying frequency[33]. Lower panel: total magnetic moment (in Bohr magneton) per Ni atom under the influence of laser pulses in the top panel. All times are in femtoseconds.

Another laser beam parameter that affects the magnetic moment is the carrier frequency of the pulse. In Fig. 4 we present the results for short laser pulses (6 fs) of peak intensity 10^{15} W/cm 2 but with varying frequency. It is clear that the central frequency of the pulse can also be used to control the amount of demagnetization. The dependence of demagnetization on frequency is non-linear and can be tuned to obtain a loss in moment of between 20% and 53% for bulk Ni. The ideal frequency needed to achieve maximum moment loss (or rather at which the system becomes most absorptive) is a material-dependent property and is related to the details of the band structure.

At this point we ask ourselves whether these three parameters can be jointly selected to get the desired demagnetization. In Fig. 5 we show results for a study. The peak intensity is kept fixed (10^{15} W/cm 2) but the frequency and duration of the pulse are tuned[34] to get

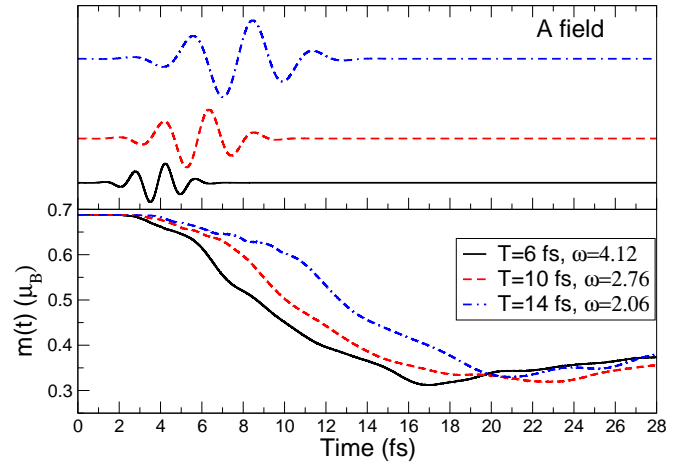


FIG. 5: (Color online) Top panel: $\mathbf{A}(t)$ of the laser pulses (see Ref. 34). Lower panel: total magnetic moment (in Bohr magneton) per Ni atom under the influence of laser pulses in the top panel. All times are in femtoseconds

the same degree of demagnetization at 20 fs. From these results we see that even though a particular set of frequency and duration combinations follow totally different paths in magnetic phase space as a function of time, at the end of 20 fs the loss in moment is the same and remains almost the same after 20 fs. It is important to mention the effect of yet another laser parameter, namely the polarization of the pulse. In the present work, linearly polarized light in x -direction was used (perpendicular to the direction of the moment, which points along the z -axis). Like experiments[35], we find that changing the plane of polarization of this linearly polarized light does not affect the process of demagnetization. In future it would be interesting to study the effect of circularly polarized light on the process of demagnetization[36]

To conclude: in the present work we show that by means of the first fully *ab-initio* TDDFT study of Fe, Co and Ni, laser induced demagnetization is a two-step process. Excitation of a fraction of electrons followed by spin-flip transitions of the remaining localized electrons is a generic process appearing in all our calculations. These results can successfully explain several experimental features like time-lag between start of pulse and demagnetization, spin-flip excitations dominating the physics initially and independence of loss of moment from the polarization of the perturbing electromagnetic field. Interestingly, we find that it is not the excited delocalized electrons which dominate the process of demagnetization but rather the remaining localized electrons. Studying the effect of easily-tunable laser parameters on the process of moment loss in Ni provides insight into optimal control of the spin moment in solids. In particular, we note that the amount of demagnetization and the time-lag between the start of the pulse and moment loss is strongly dependent on the intensity, frequency and dura-

tion of the perturbing laser pulse.

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 - [31] The pulse duration (time between the start and the end of the pulse) is 6 fs and frequency is 4.12/fs. The pulse is linearly polarized along the x -axis perpendicular to the direction of the moment. The fluence is 934.8 mJ/cm², 467.5 mJ/cm² and 93.5 mJ/cm² for the pulse of peak intensity is 10^{15} W/cm², 5×10^{14} W/cm² and 10^{14} W/cm² respectively.
 - [32] The pulse duration (time between the start and the end of the pulse) is 60 fs and frequency is 3.03/fs. The pulse is linearly polarized along the x -axis perpendicular to the direction of the moment. The fluence is 913.5 mJ/cm², 92.6 mJ/cm² and 9.3 mJ/cm² for the pulse of peak intensity is 10^{14} W/cm², 10^{13} W/cm² and 10^{12} W/cm² respectively.
 - [33] The pulse duration (time between the start and the end of the pulse) is 6 fs and the peak intensity is 10^{15} W/cm². The pulse is linearly polarized along the x -axis perpendicular to the direction of the moment. The fluence is 1009.4 mJ/cm², 934.8 mJ/cm² and 915.9 mJ/cm² for the pulse of frequency 2.06/fs, 4.14/fs and 8.24/fs respectively.
 - [34] The pulse is linearly polarized along the x -axis perpendicular to the direction of the moment and the peak intensity is 10^{15} W/cm². The fluence is 934.8 mJ/cm² for the pulse of duration 6 fs and frequency 4.12/fs, 1402.2 mJ/cm² for the pulse of duration 10 fs and frequency 2.76/fs and 1869.6 mJ/cm² for the pulse of duration 14 fs and frequency 2.06/fs.
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